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Title: **Unconventional Superconductivity and Violation of Time-Reversal Invariance**

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Unconventional Superconductivity and Violation of Time-Reversal Invariance.

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Abstract

We have discovered a low-temperature phase transition in $Bi_2Sr_2Ca(Cu_{1-x}Ni_x)_2O_8$, a high temperature superconductor. This phase transition manifests itself via a sharp drop in thermal conductivity as a function of temperature, by a factor of three within 50 mK of the transition temperature of 200 mK. The data are clear, easy to interpret and consistent with the most basic property of a superconducting transition: electrons form a condensate and drop out of the sea of normal electrons (quasiparticles) that participate in thermal conductivity. The observation of multiple superconducting phases in high temperature superconductors has been a "holy grail" of research in this field, since it would be a definitive hallmark of unconventional superconductivity. The new low-temperature state that is observed below 200 mK is expected to possess the very unusual property of violated time-reversal invariance. Our goal is to be able to differentiate between several competing scenarios for the origin of such states and their detailed nature.

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Background and Research Objectives

The nature of the mechanism that drives superconductivity in High Temperature Superconductors (HTS) has been a very active area of research since their discovery more than a decade ago. Various tools have been brought to bear on the problem, including a variety of spectroscopic, thermodynamic, and transport measurement techniques. Thermal transport has been identified as a very effective technique to investigate the nature of the superconducting state. The reason is that deep in a superconducting state electronic thermal transport is confined only to the normal electrons, since superconducting electrons (the condensate) do not carry any heat, representing the ground state of the system. This sensitivity to normal electrons in at the core of the prediction that thermal conductivity of unconventional superconductors with line of nodes of the energy gap should be linear in temperature as temperature goes to zero, and independent of the impurity concentration for a given superconductor. [1] Such universal conductivity has indeed been observed in HTS compound $YBa_2Cu_3O_7$[2], showing that the low temperature thermal transport properties of HTS support the picture of the unconventional $d_{x^2-y^2}$ superconducting order parameter in these materials. Our discovery of a second low temperature superconducting transition in Ni-doped BSCCO HTS showed on rather general grounds that HTS compounds are indeed unconventional,[3] since at least one of the superconducting states must have symmetry reduced from the one of the underlying crystallographic lattice. Moreover, the suggested $d_{x^2-y^2} + id_{xy}$ order parameter of the new superconducting state[4] possesses a property of a broken time reversal invariance, with macroscopic currents and magnetic fields appearing in a new superconducting state below the transition temperature. The purpose of this project was to use our discovery and extend the work based on it to advance understanding of the properties of both the ground state of HTS and other unconventional superconductors and its interaction with magnetic and non-magnetic impurities and magnetic field.

Importance to LANL's Science and Technology Base and National R&D Needs

Research performed under this project, both experimental and theoretical, directly enhances both the experimental and theoretical base for study of the complex materials in general, and our basic scientific understanding of unconventional superconductivity and High Temperature Superconductivity in particular. Advances in these areas will support development of technological applications of this important class of materials.

Scientific Approach and Accomplishments

Electric charge transport by quasiparticle excitations (electrical conductivity/resistivity) is shorted by the superconducting condensate that developes at high temperatures (> 70 K), and, therefore, DC and low frequency electrical conductivity is not a useful tool for probing the excitations that exist at the nodes of the superconducting order parameter. In thermal conductivity the roles are reversed: the condensate does not contribute to thermal conductivity, and one is sensitive only to the normal quasiparticle excitations. That makes thermal conductivity measurements the tool of choice for investigating the properties of normal quasiparticles deep in the superconducting state. The second (low-temperature) superconducting transition directly affects the spectrum of normal quasiparticles and thereby dramatically changes the thermal conductivity of a sample, as shown in Figure 1. However, electrons are not the only carriers of heat in the solid. Phonons (lattice vibrations) also carry thermal current.

In order to firmly establish that the 200 mK anomaly in thermal conductivity is due to electrons, we must separate these two channels. In order to identify the phonon contribution to thermal transport, we have extended thermal conductivity experiments to measurements of an insulating analogue $B_{i2}Sr_2(Y_xCa_{1-x})Cu_2O_8$, where substitution of yttrium for calcium drives the sample from superconducting to insulating ground state.[5] The resulting thermal conductivity of insulating samples is about an order of magnitude below that of superconduting samples (see Figure 1). This results allowed us to show that the 200-mK anomaly is indeed of electronic origin. We also have performed additional higher temperature thermal conductivity studies on insulating $B_{i2}Sr_2(Y_xCa_{1-x})Cu_2O_8$, which proved the high quality of the samples compared to those investigated and reported by other researchers.

We showed theoretically that the $d_{x^2-y^2}$-wave superconductor is marginally stable in the presence of external perturbations. Subjected to the external perturbations by magnetic impurities, it develops a secondary id_{xy} component of the gap, to maximize the coupling to impurities and lower the total energy of the state. The secondary id_{xy} component exists at high temperatures and produces the full gap ≈20 K in the single particle spectrum around each impurity, apart from impurity induced broadening. At low temperatures the phase ordering transition into global $d_{x^2-y^2}+id_{xy}$ state occurs.[3] We have generalized the notion of the marginal stability of the $d_{x^2-y^2}$-wave superconductor to a variety of external perturbations, including magnetic field. In the presence of a magnetic field the compound upon cooling from normal state goes first directly into a $d_{x^2-y^2}+id_{xy}$ state as shown in Figure 2. This discovery carries potential significance to the application of the HTS compounds in various devices that either create magnetic field or operate in a magnetic field environment. Therefore, understanding of such time reversal invariance states is important for technological implications.

To investigate experimentally the additional effects of a magnetic field on superconducting state of HTS compounds we performed preliminary thermal conductivity measurements on a pure supercomputing Bi2212 sample in a magnetic field (H) applied parallel to the CuO_2 layers responsible for high-temperature superconductivity. In this orientation, only the magnetic spin of superconducting electrons couple to magnetic field, since the magnetic vortices penetrating the sample lie between the superconducting planes and do not lead to additional supercurrent. Figure 3 shows linear dependence of the increased thermal conductivity on magnetic field, contrary to the predicted H^2 dependence. To resolve this anomalous observation, we have acquired a precision rotator, which will be installed in a dilution refrigerator capable of achieving temperature of 20 mK. Magnetic field of up to 9 Tesla at the rotator will be provided by a superconducting magnet. The detailed study of the field dependence of thermal conductivity on the angle between the field and the CuO planes will provide additional tests of the two-dimensionality of HTS.

We used thermal conductivity techniques developed within the course of this project to study a new family of unconventional superconductors: heavy fermion $CeIrIn_5$ (T_c = 0.4 K) and $CeCoIn_5$ (T_c = 2.3 K).[6] We were able to observe the predicted universal thermal conductivity limit (as T--> 0 K) (where thermal conductivity is independent of the amount of impurities present in the sample) in $CeIrIn_5$ (Fig. 4). Such behavior is only expected in unconventional superconductors with lines of nodes in the energy gap, as in two-dimensional, d-wave, high-temperature superconductors. Thermal conductivity in $CeCoIn_5$ displays a kink and a sharp rise as the sample becomes superconducting at T_c = 2.3 K (Fig. 5). Such behavior closely parallels that observed in a number of HTS. It is generally attributed to the strongly reduced scattering of the electrons in the superconducting state, due to decrease in antiferromagnetic fluctuations. Observed behavior in $CeCoIn_5$ hints at the importance of magnetic fluctuations for unconventional superconductivity in heavy fermion compounds. In addition, we observe a close to T^3 behavior of thermal conductivity in $CeCoIn_5$ at very low temperatures of $T/T_c < 0.043$. Such behavior is expected in the clean limit of unconventional superconductors with line of nodes in energy gap.

To confirm this picture we performed low-temperature specific heat C measurements of both $CeIrIn_5$ and $CeCoIn_5$. Both compounds show the presence of linear-in-T and T^2 terms in C, indicating the presence of the nodes in energy gap, in accord with thermal conductivity measurements.

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11. R. Movshovich, M. Jaime, J. D. Thompson, C. Petrovic, Z. Fisk, P. G. Pagliuso, and J. L. Sarrao, "Unconventional superconductivity in $CeIrIn_5$ and $CeCoIn_5$: Specific heat and thermal conductivity studies", submitted to *Phys. Rev. Lett.*

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6. R. Movshovich, M. Jaime, J. D. Thompson, C. Petrovic, Z. Fisk, P. G. Pagliuso, and J. L. Sarrao, "Unconventional superconductivity in $CeIrIn_5$ and $CeCoIn_5$: Specific heat and thermal conductivity studies", submitted to *Phys. Rev. Lett.*

Figures

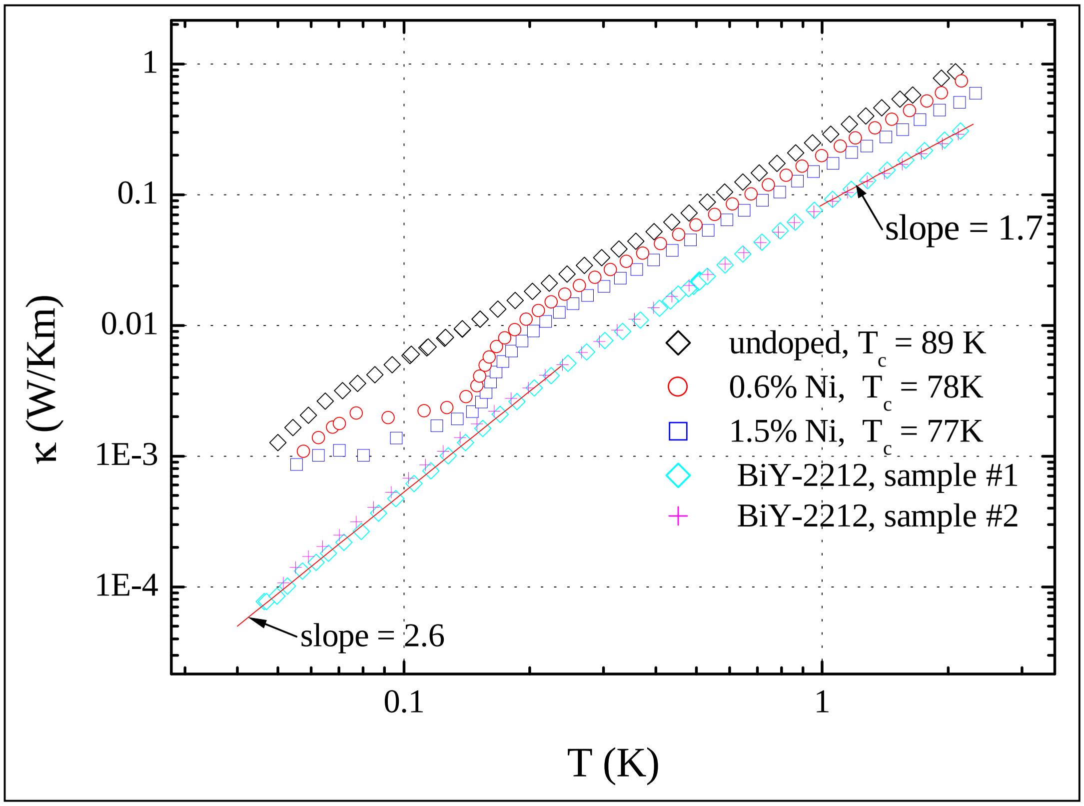

Figure 1: Thermal conductivity of several superconducting BSCCO samples and its insulating analogue BY-2212, Y-doped BSCCO samples. Insulating samples clearly have much lower conductivity than superconducting ones, suggesting that at very low temperature most of the heat in superconducting samples is carried by electrons

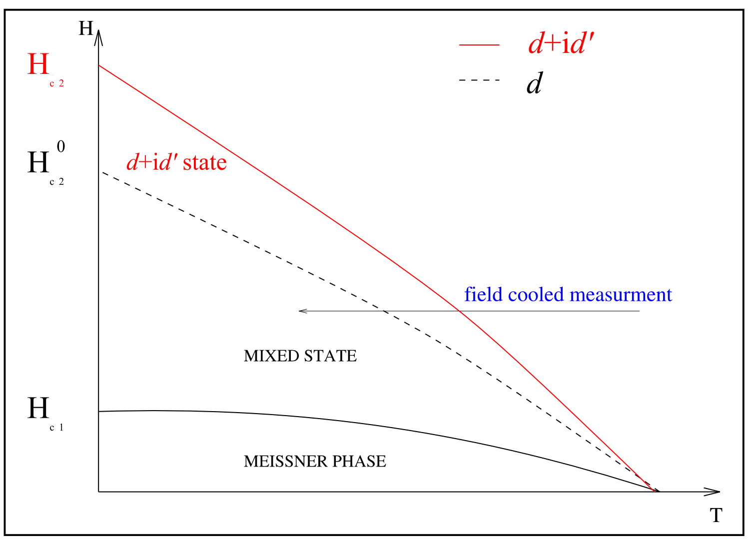

Figure 2: The (H,T) phase diagram of d-wave superconductor. Magnetic field leads to introduction of the $d_{x^2-y^2}+id_{xy}$ state above the usual mixed (vortex) state.

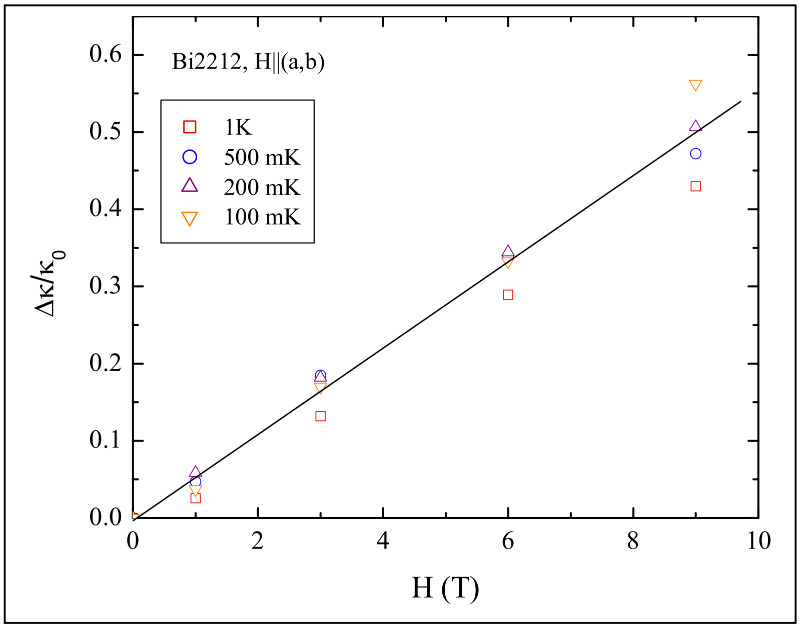

Figure 3: Relative increase of thermal conductivity of superconducting BSCCO in a magnetic field applied in the CuO_2 planes. Linear dependence on the field is observed.

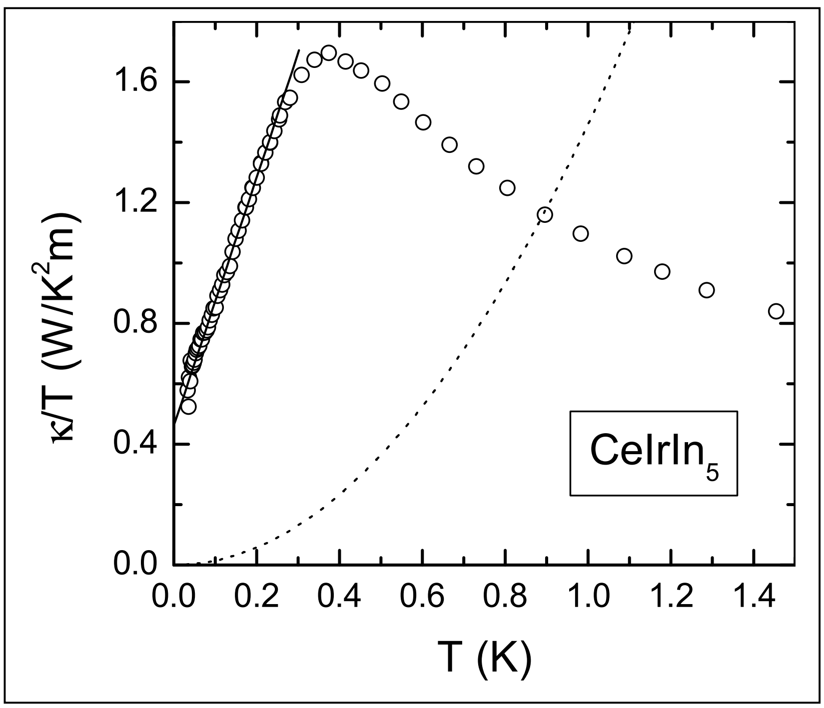

Figure 4. Thermal conductivity of $CeIrIn_5$. Solid line is a linear fit to the data for T < 0.2 K = $T_c/2$. Dotted line is an upper limit estimate for the phonon thermal conductivity.

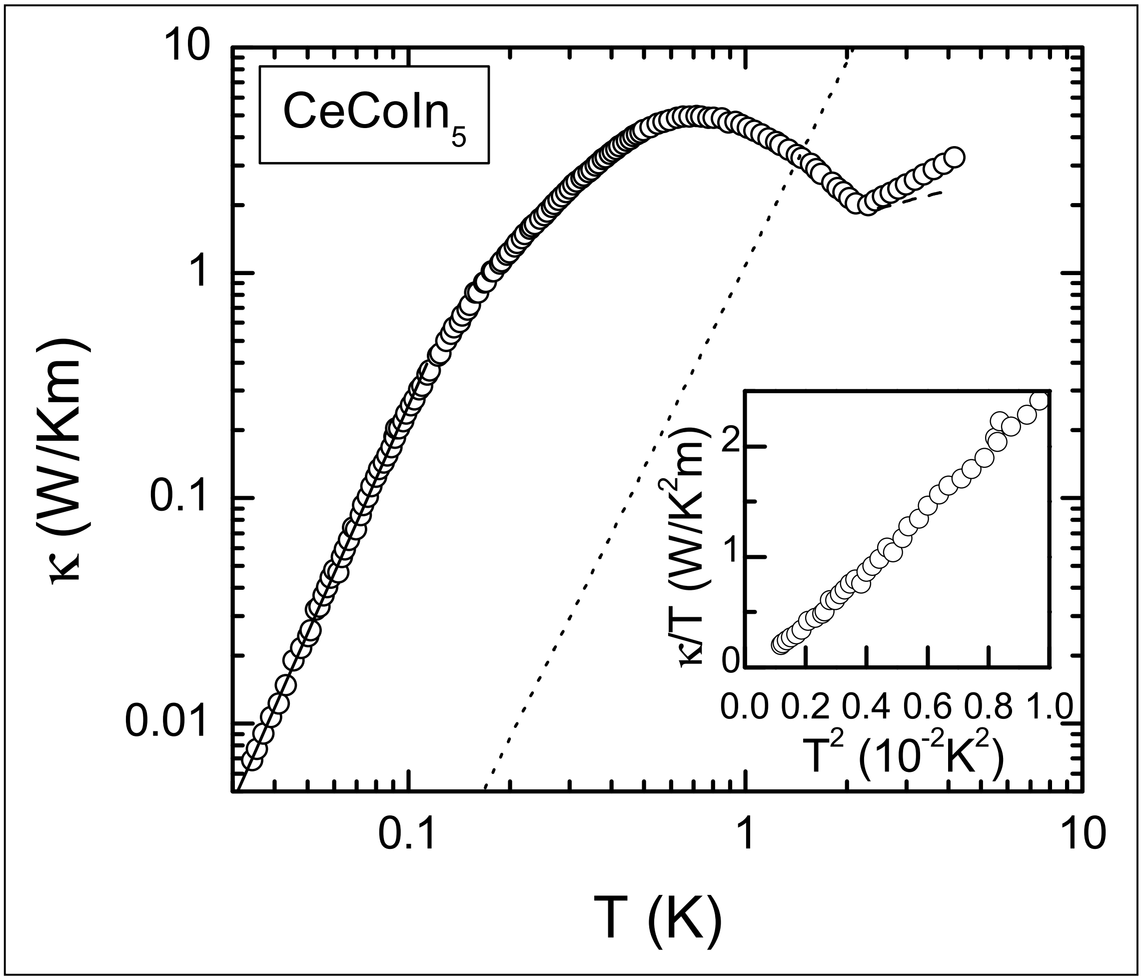

Figure 5. Thermal conductivity of $CeCoIn_5$. Solid line is a power law fit for T < 100 mK < $T_c/20$. Dotted line is an upper limit estimate of phonon thermal conductivity. Dashed line is electronic thermal conductivity for T > T_c. Inset: thermal conductivity over temperature vs. T^2 for T < 0.1 K.